ASSESSMENT OF A RADIOACTIVE WASTE DISPOSAL SITE AT ENEWETAK ATOLL

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Abstract—The 43 nuclear tests conducted at Enewetak Atoll by the United States between 1948 and 1958 produced close-in fallout that contaminated the islands and lagoon of the atoll with radioactive fission and activation products, and unfissioned nuclear fuel. In 1972, the U.S. government announced that it would conduct a cleanup and restoration operation to return the atoll to the Enewetak people. The radiological cleanup began in 1977 and lasted to 1980 and focused on reducing the concentration of the transuranium elements $(^{238,239,240}$ Pu and 241 Am = TRU) in soils on some of the islands that might eventually be used for residence or for subsistence agricultural. The cleanup plan called for relocating soil and some other contaminated debris to Runit Island on the eastern perimeter of the Atoll. Some of the contaminated soil was mixed with cement and the mixture placed below the water level in the Cactus Crater that was formed by a nuclear explosion in 1958. The remainder of the contaminated material was mixed with concrete and placed above ground over the crater in the shape of a dome. A concrete cap was constructed over the dome of soil. Concern has been expressed by the people of Enewetak and by others over the possible aquatic impacts from the radionuclides entombed in the crater. A National Academy of Sciences committee examined the dome and concluded that the containment structure and its contents present no credible health hazard to the people of Enewetak, either now or in the future. The committee suggested that "at least part of the radioactivity contained in the structure is available for transport to the groundwater and subsequently to the lagoon and it is important to determine whether this pathway may be a significant one." Therefore, a surveillance program was started in 1980, in conjunction with other research efforts, to study the radionuclides in samples of fish, groundwater, and lagoon seawater. Our data and conclusions support the findings suggested by the National Academy committee over a decade ago in that any assumption of rapid remobilization of all or any of the dome's transuranics or other radionuclides is an extreme one. Any fear that this structure contains amounts of activity whose release would cause damage to the environment that will result in greater effect on human health is unfounded.

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INTRODUCTION

The 43 nuclear tests conducted at Enewetak Atoll by the United States between 1948 and 1958 produced close-in fallout that contaminated the islands and lagoon of the atoll with different amounts of radioactive fission products, activated products, and unfissioned nuclear fuel. Quantities of concrete, metal debris, cable, bunkers, buildings and other miscellaneous materials, some contaminated and some not, were also abandoned at the Atoll after the U.S. testing program finished. In addition there were U.S. non-nuclear programs between the years 1958 and 1972 that also modified the landscape on some islands of the Atoll.

Enewetak Atoll is located in the Equatorial Pacific Ocean at approximately 11°21′N and 162°21′E in the northwestern portion of the Republic of the Marshall Islands. The islands of the Atoll are shown in Fig. 1 along with the location of the major nuclear craters. The Marshallese name for each island and two large coral heads are also shown. U.S.-assigned names for the islands during the testing period are given in parenthesis.

In 1972, the U.S. government announced that it would conduct a cleanup and restoration operation to return the atoll to the Enewetak people. Planning for the cleanup extended from 1972 to 1977. The final project was conducted as a series of concurrent tasks between May 1977 and April 1979. It involved several departments of the federal government with the Defense Nuclear Agency (DNA) responsible for cleanup activities. The radiological cleanup concentrated on reducing the surface soil levels of the transuranium elements $(^{238,239,240}$ Pu and 241 Am = TRU) on some of the islands that might eventually be used for residence or the growing of subsistence agricultural products. The justification for basing the cleanup on transuranic criteria can be found in the description of planning efforts, cleanup operations, and radiological guidelines in Defense Nuclear Agency and Department of Energy documents (U.S. DNA 1981; U.S. DOE 1982). Other miscellaneous debris and radioactive material from test-day burial sites were also identified for removal. Only the quantities of transuranics were measured during field operations, but soil relocation also involved moving undetermined amounts of long-lived fission and activation products associated with the carbonate soil.

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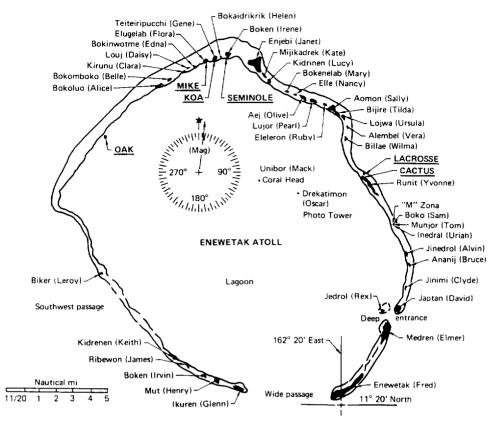


Fig. 1. Islands of Enewetak Atoll with Marshallese and English code names. Locations of major nuclear craters are indicated.

The cleanup plan called for collecting and transporting the soil and other contaminated debris to Runit Island on the eastern perimeter of the Atoll. The contaminated soil was mixed with cement and the mixture placed below the water level (tremie method) in the Cactus Crater that formed during a 1958 nuclear explosion on the northern tip of the Runit reef. The 10-m-deep crater was filled to the low-tide level by this method. Above the water level, the contaminated soil was blended with cement using a disc-harrow. Water was applied and the mixture was compacted. Some solid objects were added to the above ground slurry. A dome-shaped mound of contaminated material was thereby formed over the crater. A central "donut" hole was left in the center of the dome and eventually filled with soil and debris removed from other locations on Runit Island. The mound was finally covered with concrete panels to form a cap over the contents. The crater was surrounded by a concrete key wall to reduce scouring and undermining by wave action. A report on the radiological cleanup of Enewetak Atoll (U.S. DNA 1981) provides a detailed account on the construction and filling of the dome during cleanup.

The structure is referred to as a waste disposal site since it covers material contaminated with quantities of long-lived radionuclides. There have been many expressions of concern by the people of Enewetak and by others over the possible aquatic impacts from the radionuclides entombed in this disposal site. The people are convinced that it must be one of the most dangerous places at the Atoll since the United States spent millions of dollars to contain radioactive material under the domed structure. It is imagined that if leakage were to occur, many marine resources would be affected. News segments recorded on film and shown on television in the 1980's helped to establish a fear and concern about this disposal site.

In 1980, the Defense Nuclear Agency requested the National Research Council of the U.S. National Academy of Sciences to establish a committee of experts to evaluate the "effectiveness of the Cactus Crater structure in preventing harmful amounts of radioactivity from becoming available for internal or external human exposure." In a report published in 1982 (NAS 1982), the committee concluded that the Cactus Crater containment structure and its contents present no credible health hazard to the people of Enewetak, either now or in the future. However, there were issues related to the permeability of the tremie concrete. There were sections in the tremie that were in free communication with the ocean. This led the committee to suggest that "at least part of the radioactivity contained in the structure is available for transport to the groundwater and subsequently to the lagoon and it is important to determine whether this pathway may be a significant one." Therefore we initiated an environmental surveillance program in 1980 to study the radionuclides in samples from the vicinity of the dome. This included the collection, processing and analysis of samples of fish, groundwater, and lagoon seawater. Samples were taken until 1984 when the program was terminated.

Subsequent to the cleanup there were data from comparable samples collected from Runit and other islands of the Atoll in support of other research activities. Comparison of the pre- and post-cleanup data indicated that there was no adverse radiological impact on the environment from the radionuclides contained in the Cactus crater structure. All aquatic data generated during the period appeared in U.S. Department of Energy progress reports and has not been previously published outside this report. During the 1990's, we again collected samples to assess what, if any, changes in radionuclide concentrations occurred during the intervening years. This document examines the present and past concentrations of plutonium and other radionuclides at the Atoll and in the immediate environment of the disposal site. Published and unpublished concentrations measured in aquatic environmental samples (seawater, sediment, species of fish, groundwater) collected before filling the crater are compared with levels in corresponding samples collected after the crater was filled. The concentration of plutonium and other radionuclides measured in the material filling the crater and found above ground level under the dome is summarized. Implications of all results that relate to the disposal site on Runit Island are discussed.

METHODS

The majority of results from samples discussed in this report were generated over the last three decades. Field and laboratory personnel changed during this period, but collection methods, sample processing, and radiochemical procedures did not significantly change. Therefore, previously published documents adequately describe field collections in the Marshall Islands and the analytical techniques used at Lawrence Livermore National Laboratory for analysis and data reduction. Collection and processing sea and ground-water may be found in Marsh et al. (1975); Noshkin et al. (1976); Marsh et al. (1978); Noshkin et al. (1981b); Noshkin et al. (1974); and Noshkin et al. (1987). Collection, description, and analysis of parts of different species of fish are found in Noshkin et al. (1981a) and Noshkin et al. (1988). Sediment collection, processing, and analysis are given in Nelson and Noshkin (1973) and Noshkin (1980). It will not be possible to list the many results discussed in this report. Only summaries of available data will be shown in the Tables and Figures appearing in this document.

RESULTS AND DISCUSSION

Radiological conditions Runit Island prior to cleanup

Runit (Yvonne) Island, identified in Fig. 1, is located on the eastern perimeter of the Atoll. The island and the adjoining reef were used for several nuclear tests (4-surface; 5-tower; 1-atmospheric), and 8 more devices were detonated on barges located off-shore of the island in the lagoon. It was the most severely radiologically contaminated island at the atoll. Barges for seven of the tests were anchored in the lagoon from 170 to 1,200 m offshore the island. During the 1958 Quince test on Runit, only the high explosive component of the device was detonated. This resulted in scattering the plutonium nuclear fuel over a large area of the island. To prepare for the Fig event, scheduled 12 d later in the same location, 3 to 5 inches of the plutonium contaminated soil was bulldozed from the site and disposed of in the lagoon immediately offshore the center of the island (U.S. DNA) 1981; U.S. DOE 1982). The transuranics resulting from the barge events and the bulldozing operations were identified in the near shore sediments and quantified during a 1972 radiological survey (Nelson and Noshkin 1973). These results show that the mean quantity of TRU's distributed over the surface sediments (to a depth only of 2.5 cm) in a 0.7 km² region extending 0.8 km lagoonward of the island is about 64 GBq. The mean concentration and inventory of the transuranics in the surface sediment offshore Runit and in the entire lagoon (Nelson and Noshkin 1973; Noshkin 1980) is summarized in Table 1. The lagoon sediment contains the largest reservoir of plutonium at the Atoll. These sediments are exposed to the bottom waters of the lagoon and the radionuclides are remobilized continuously to the hydrosphere from the sedimentary source term. Mean water concentrations of plutonium measured in the lagoon over time are shown in Table 1. These data demonstrate that remobilization is occurring from the sediments to maintain an inventory of plutonium in the lagoon water mass decades after testing. At Runit, the plutonium in the near shore sediments is also mobilized and measured in seawater and has been available for uptake by near shore organisms for many years (Nelson and Noshkin 1973; Noshkin et al. 1974; Noshkin et al. 1976; Noshkin 1980; Noshkin et al. 1981a).

Radiological conditions at Cactus Crater subsequent to cleanup

Cactus Crater was formed in May 1958 by the 18-Kt Cactus event detonated on a manmade extension of Runit island on the lagoon side of the reef. Cactus Crater in the foreground and LaCrosse Crater with the ocean in the background are shown in Fig. 2a at a time before cleanup. The test produced a crater roughly 112 m in diameter and 10 m deep. When the device exploded, some of the pulverized material fell back into the crater so that the original hole was deeper than 10 m. Quantities of different radionuclides are distributed non-uniformly throughout the sediment sampled in the 10–15-m-thick

Table 1. Transuranic radionuclide lagoon sediment and plutonium water column inventories. Sediment and water data from Noshkin (1980); Noshkin et al. (1987).

Sediment concentrations				
Enewetak Atoll (area, 933 km²)	239 + 240Pu	^{2,38} Pu	²⁴¹ Am	Total TRU
Areal activity to 2.5 cm depth (GBq km ⁻²)	9.9	1.4	3.0	14.3
Total to 2.5 cm depth (GBq)	9,200	1,300	2,800	3,300
Total to 16 cm depth (GBq) ^a	44,000	6,200	17,600	67.800
Region to 0.7 km offshore Runit (0.86 km ²)				
A real activity to 2.5 cm depth (GBq km ⁻²)	56.5	14.1	5.7	76.3
Total to 2.5 cm depth (GBq)	47.5	11.9	4.8	64.2
Total to 16 cm depth (GBq) ^a	225	57	30	312

Average ²³⁹⁺²⁴⁰Pu lagoon water concentration (Bq m ⁻³)/inventory (GBq) (Enewetak Lagoon area 933 km²; mean depth 0.049 km)

Date of collection	Number of samples	Soluble	Concentration particulate	Total	Total inventory
October-December 1972	35	0.81	0.3	1.18	53.9
July 1974	71	0.93	0.70	1.63	74.5
May 1976	29	0.59	0.48	1.07	48.9
May 1982	23	0.63		c	

^a Core samples collected over the lagoon in 1972 and 1977 showed only 21 \pm 11% of the plutonium and 16 \pm 6% of ²⁴¹Am in the sediment column are associated with the surface 2.5 cm surface layer. These values are assumed representative for sediment anywhere in the lagoon to estimate inventories to 16 cm from measured surface concentrations (Noshkin 1980). ^b Total transuranics (238 Pu + $^{239+240}$ Pu + 241 Am)

^e Particulate phase not measured.



Fig. 2a. Cactus crater in foreground with LaCrosse crater and the ocean reef in the background at a time before cleanup activities.

fallback zone of altered carbonate material (Ristvet et al. 1978). The crater resembled a spherical segment with a flat base and had an average volume at mean sea level of $3.3 \times 104 \text{ m}^3$ but could hold up to $4.4 \times 104 \text{ m}^3$ of water during periods of highest tide. The total surface area is approximately 6,900 m², of which only an estimated

2,060 m² were covered with sedimentary deposits. The remaining upper slopes were littered with rock rubble although some areas of the wall did have thin veneers of coarse sand. Much of the surrounding rock is heavily fissured from events detonated nearby. The majority of the crater rim is on land, but about a quarter of the eastern circumference was open to permit exchange of water between the crater and ocean during periods of high tide. Circulation of seawater in the region is directly affected by the windward cross-reef currents.

The groundwater in the area immediately southeast of the crater flows generally southwest into the lagoon (Noshkin et al. 1976). Dye tracer studies (Marsh et al. 1978) showed that most of the water lost from the crater is by overflow during periods of high tide. The water eventually flows into the lagoon through a break in the land extension some 400 m northwest of the crater (Noshkin 1980; Marsh et al. 1978). The dye studies also showed that only small amounts of crater water enter the island's groundwater or flow subterraneanly into the lagoon. The residence time of the water in the crater was a function of the tidal range and could be predicted for any period with available tide data. The mean residence time of the water, averaged over a month, was about 2.6 d (Marsh et al. 1978).

Plutonium and other radionuclides were supplied to the crater water by three processes: transportation of some quantities were associated with surface ocean water advecting over the reef; release to the bottom interstitial water occurred from the contaminated bottom sediments of the fallback zone; and by interactions involving resuspended bottom sediments with the crater water.

The latter two mechanisms contributed most of the plutonium radionuclides to the erater water column. Between January 1975 and May 1977, 27 seawater samples from different depths in the crater were obtained for radionuclide analysis. Table 2 summarizes the mean concentrations for ²³⁹⁺²⁴⁰Pu, ¹³⁷Cs, ⁹⁰Sr and S values [amount of ²³⁸Pu to total plutonium (²³⁸⁺²³⁹⁺²⁴⁰Pu) alpha activity] in filtered water and particulates from depths in the crater. Filtered interstitial water and short sediment cores were sampled between 1974 and 1977. Concentrations in these samples are also shown in Table 2. With the estimated exchange rate and size of the plutonium sediment reservoir it is estimated that 11.5 MBq of ^{239 + 240}Pu and approximately half this amount of ²³⁸Pu is annually released from the crater bottom sediments. This plutonium mixes with the seawater along the reef and subsequently merges with the inventory contained in the lagoon water mass. The crater source contributed about 0.03% to the annual average lagoon water (soluble) inventory of ²³⁹⁺²⁴⁰Pu (see Table 1). Filling the crater with solid debris and closing the access of ocean water on the eastern perimeter during cleanup was an effective means of eliminating this small contri-

Table 2. Concentrations of radionuclides in cactus crater water, sediment, and interstitial water samples collected between February 1974 and May 1977 prior to cleanup activities.

	Solu	tion	Partic		Solution	Particulate	Solution
	^{2,39+240} Pu	S Value ^a	^{239 + 240} Pu	S Value ^a	137Cs	²⁴³ Am	⁹⁰ Sr
Mean surface water	2.6 ± 0.7	0.32	4.0 ± 2.1	0.33	8.9 ± 2.6	<1.5	
Mean mid-depth (4.5m)	3.2 ± 1.1	0.33	4.2 ± 1.3	0.33	8.1 ± 1.8		
Mean bottom (9 m) water	3.6 ± 1.7	0.33	16.1 ± 8.6	0.33	8.9 ± 2.5		
Mean crater water	3.2 ± 0.9	0.33	8.3 ± 5.1	0.33	8.6 ± 0.6	<]	-17.0 ± 3.0
(27 samples for plutonium)							

Concentration of ²³⁹⁺²⁴⁰Pu and ¹³⁷Cs in interstitial sediment pore water (Bq m⁻³).

	236	· 240Pu	SV alue	¹³⁷ Cs
29 May 1977 Outgoing tide		8.8 ± 0.4	0.35	10.1 ± 1.3
Outgoing tide		8.5 ± 0.5	0.34	8.3 ± 1.1
30 May 1977 Low tide		12.3 ± 0.6	0.34	7.7 ± 0.4
	Average	9.9 ± 2.1	0.34	8.7 ± 1.2

Concentration of some radionuclides in crater bottom sediment (Bq g⁻¹ dry wt.)

	^{239 + 240} Pu	SValue	^{-137}Cs $^{-24}$	ⁱ Am
Collected 2/2/74				
0-5.7 cm surface section	3.04 ± 0.07	0.35	0.46 ± 0.04	0.34 ± 0.07
5.7-11.4 cm section	2.93 ± 0.19	0.35	0.52 ± 0.04	0.39 ± 0.07
11.4–17.2 cm section	3.74 ± 0.26	0.35	0.56 ± 0.04	$1 - 0.39 \pm 0.07$
Collected 4/26/76				
0-2 cm surface fine fraction	3.85 ± 0.15	0.34		
0-2 cm surface coarse fraction	1.38 ± 0.12	0.33		
4–6 cm fine fraction section	3.26 ± 0.32	0.32		
4-6 cm coarse fraction section	2.46 ± 0.25	0.36		
10–14 cm fine fraction	4.06 ± 0.32	0.35		
10-14 cm coarse fraction section	2.82 ± 0.28	0.39	10 1 miles 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	·

^a S Value. ²³⁸Pu to total plutonium alpha activity ratio.

bution of plutonium and other radionuclides to the lagoon water.

The cleanup

The cleanup of Enewetak extended from May 1977 to April 1980. Island surveys were made with in-situ monitoring equipment to measure surface levels (to a depth of 3-5 cm) of ²⁴¹Am over an established grid on each island. Soil samples were obtained for laboratory analysis of plutonium radionuclides and americium to develop ratios between the TRU's and ²⁴¹Am. These analyses provided the data to develop radiological contour maps that were used by crews during cleanup activities. After sections of islands were identified for cleanup, solid rubbish was removed followed by the removal of several centimeters of topsoil. The soil was loaded onto barges and transported to Runit where it was off-loaded in a stockpile near the crater. The soil was filtered through a 3.8-cm screen to remove oversized particles, mixed with cement and attapulgite to form a mixture designed for use in the "tremie" method of placing a concrete mixture under water. A concrete pump transferred the slurry through a pipe to the bottom of the crater, displacing the overlying water. A number of problems were encountered during the tremie operation. but in the end some 41,600 m³ of soil filled the crater to the low-tide water level using this method. Above the water level the soil was blended with cement using a disc-harrow, and each layer was compacted. Following this procedure a dome-shaped mound of soil was formed over the crater. This material is not in communication

with the subterranean groundwater. A central "donut" hole was left in the soil dome. This space was reserved primarily for debris translocated from other parts of Runit Island. After the hole was filled with soil and other debris, the 46-cm-thick concrete cap or dome was completed. A view taken over the lagoon of the dome in the final stages of the cleanup operation is shown in Fig. 2b with LaCrosse Crater and the ocean reef in the background.

The amount of soil and TRU's transferred both to the crater and placed above ground under the dome are abstracted from clean up records (U.S. DNA 1981) and shown in Table 3. Table 3 also provides the quantity of soil and TRU's removed from the different islands. Only 24% of the total TRU was buried below ground level in the crater while the remaining activity is associated with material placed above the water level under the dome. Using a dry weight soil density of 1.29 g cm⁻³ (U.S. DOE 1982), the average soil TRU concentration in the undiluted crater fill is computed from the data in Table 3 to be 2.4 Bq g⁻¹. Table 2 shows that the total TRU in the surface 17 cm of bottom sediment from the crater sampled in 1974 was about 4.7 Bq g⁻¹. This is nearly twice the concentration in the material used to fill the crater. Therefore, if leakage were to occur into the groundwater from the fill, it would be difficult to detect. There should be less TRU mobilized to solution from the fill than was previously mobilized from the crater bottom sediments and found in the crater water (shown in Table 2) before 1977.

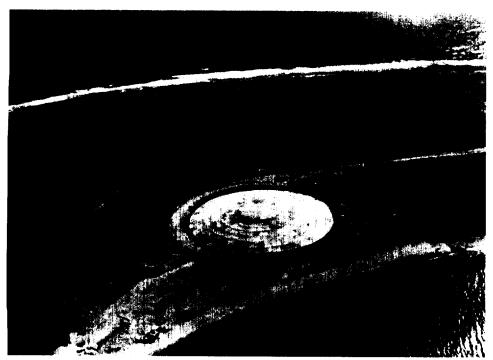


Fig. 2b. View from over the lagoon of the Cactus dome in the final stages of completion. LaCrosse crater and ocean reef in the background.

Table 3. TRU activity and volume of soil excised and placed in Cactus Crater and under the dome on Runit Island.^a

	TRU Activity	Soil remov	ved (m ³) to
Island	GBq	Crater	Dome
Aomon (Sally)	48.1	8,100	0
Aomon Crypt (Sally)	33.3	342	7,130
Boken (Irene)	37.0	322	3,450
Enjebi (Janet)	96.2	32,890	7,633
Lujor (Pearl)	63.0	0	11,415
Runit (Yvonne)	267.4	0	8,210 ^b
Total	545°	41,654	37,838

a Data from DNA 1981.

Approximately one-half of the total inventory of TRU now under the concrete dome originated from only 5 northern islands. The remaining material was surface material translocated from one or more areas on Runit and dumped above ground in the crater donut hole. The amount of activity moved to Runit from the 5 northern islands is comparable to the inventory of TRU already in the lagoon sediments to a depth of 16 cm in the 0.86 km² area off-shore the island (see Table 1). Comparison of values in Tables 1 and 3 also shows that the inventory of the TRU's in this waste disposal site is equivalent to only 0.8% of the total TRU inventory in the lagoon sediment to a depth of 16 cm. Therefore, if the contents were to find its way into the lagoon, the inventory of the TRU's in the local area, and especially in the entire lagoon, would not change by any significant amount and no unacceptable hazard would result (NAS 1982) if such a catastrophic event was ever to occur.

Results from the 1980 National Academy study

In March 1980, members of the National Academy of Sciences committee visited the Atoll to conduct a series of sampling and observations at the dome. Tests included taking solid core samples from holes drilled through the soil-cement and tremie fill. Three holes were drilled through the third concrete ring from the top. Material from the center "donut" hole was not sampled during these tests. Water samples were also taken from two different levels in one drill hole from below ground level in the zone of incompletely cemented tremie concrete. In the tremie zone there was relatively free communication with the groundwater (NAS 1982). The soil-cement above the water level also did not achieve the concrete-like character that was anticipated. The cores from the bore holes were sectioned and the material from different depth intervals was described. Sections were then analyzed by gamma spectrometry, and the transuranics and 90Sr were determined following radiochemical separation (Robison and Noshkin 1981). Table 4 shows a description of the material encountered at depths in the different zones of the structure along with the concentration of ²³⁹⁺²⁴⁰Pu measured in sections of the 3 cores. Concentrations of the radionuclides measured in the filtered water samples and the particulates are shown in Table 5. The description in Table 4 shows there is poorly cemented soil in both regions under the dome, and there also appears to be considerable amounts of debris in the fill, other than soil. Table 6 lists the mean concentrations for the principle radionuclides measured in the samples from the tremie section below ground and from the soil-cement region above ground under the dome. These mean values are developed from the core data in Robison and Noshkin (1981). The mean concentration of the TRU shown in Table 6 is approximately 3–4 times less than the soil concentration expected from the results shown in Table 3. However, this difference should not be considered unreasonable since the estimates of radionuclide concentrations made during the cleanup are not likely to be very accurate (NAS 1982), and the dilution of the soil concentrations with the uncontaminated cement (estimated to be about 30% of the measured value) was not considered.

It is probable that the measured concentrations from the Academy samples more accurately represent the TRU's in the material below ground in the crater and above ground, exclusive of the contents in the center hole, than the estimates made from the field measurements (U.S. DNA 1981) during the cleanup. Based on these measured values, the mean TRU inventory below ground would be 34 GBq and above ground the inventory would be 50 GBq, exclusive of the amount associated with the fill of the center hole. Interestingly, the mean concentrations of 0.62 Bq g^{-1} and 1.3 Bq g^{-1} , respectively, in the crater and dome soil, shown in Table 3, are less than the post cleanup average TRU levels (U.S. DOE 1982) measured in surface soils (1–3 Bq g⁻¹) on many of the northern islands such as Alice, Belle, Clara, or Daisy (see Fig. 1). These islands satisfy cleanup criteria where the Enewetak people can visit and gather food.

The average concentration of ²³⁹⁺²⁴⁰Pu found in solution within the containment structure is approximately one-half the value found in the crater seawater before the crater was filled with debris. Comparing the soluble water concentrations in Table 5 with the levels in water from Table 2 shows that the fill lowered the concentration of plutonium (and TRU) but also caused a significant increase in the dissolved concentrations of ¹³⁷Cs and ⁹⁰Sr. Therefore, if leakage of crater material were to occur through the groundwater aquifer, ¹³⁷Cs would be a better tracer for this source of water since it is released to and moves in solution more readily than the TRU's.

Concentration of radionuclides in samples from the surrounding environment of North Runit

Groundwater and off-shore seawater. A ground-water program was initiated at Enewetak Atoll in 1974 to study the hydrology and groundwater geochemistry on selected islands of the Atoll including Runit. Groundwater from 2 well sites (XRU 5 and XRU 6) between the

^b Most activity and soil in central donut hole under dome.

^e TRU estimated in Crater 131 GBq.

^{*} TRU estimated in Dome 147 GBq without amount in Donut hole and 414 GBq including amount in Donut Hole.

Table 4. Description of material and plutonium in samples from interior sections of the dome.

Depth (m)		Description of	Mean ²³⁹⁺²⁴⁰ Pu (Bq g ⁻¹) in extracted soil sample ^b drill hole		
Under	Dome	sample material ^a	CD-I	CD-12	CD-17
0	0				
	0.43	Dome cap			
1					
2		Uncemented medium-	1.41	1 22	1.70
2		fine soil-cement		1.22	<u>1.70</u>
3		me son-cemen	0.37		
2.			<u>0.57</u>		
4	3.8			1.37	0.74
		Oversize material			
5	5.3	cobble, limbs, wire, rebar			
	5.2		dome fill a below	bove this depth an	d crater fill
6		0.15-m layers of			
		poorly cemented tremie	0.06		
7	6.7	with uncemented soil		0.19	
		Oversize debris and cobble			
8			0.44	0.23	0.55
9		Layers of poor to moderate cemented tremie and soil, cobble, rebar, wood	0.78		
10	9.8			0.28	$\frac{-}{0.41}$
		Soil, wood, minor gravel-			
11		size tremie			
	11.3			0.005	0.59
12		Fallback material below this depth.	Original crater sed	iment	
13				0.056	
			0.41	0.000	
14					
				0.008	
15					
16	15.8			0.008	
		Bottom original crater			

^a Description of material in core hole CD-1 as provided in NAS (1982). Radiological data from Robison and Noshkin (1981).

^b Measured concentration is indicated within depth interval sampled.

crater and the lagoon were regularly sampled between 1974 and 1979. Some of our early results on radionuclide concentrations in Runit groundwater have been discussed previously (Noshkin et al. 1976). The well sites were destroyed during cleanup operations sometime between April and October 1979, but they were replaced by DNA in March 1980. The two new wells were identified as CW1 and CW2. CW1 is approximately 5 m from the base of the dome near where XRU 5 was located, and CW2 is 15 m lagoonward of the base near the site of XRU 6. Since there was a small but perceptible groundwater flow in the region towards the lagoon, it was felt important to monitor the surface water for any changes in radionuclide concentration. Water was sampled during

trips to the Atoll from 1980 to 1984 and again during the 1990's. Average concentrations of ²³⁹⁺²⁴⁰Pu and ¹³⁷Cs in groundwater during periods of pre- and post-cleanup are shown in Table 7. ¹³⁷Cs measurements are included because it is more mobile than plutonium and is therefore a better indicator of any leakage from the crater site.

Seawater was sampled from the lagoon 50–100 m opposite well CW2 in approximately 8 m of water. We designate this location as station CL-1. Station Cl-1 was the location where the research vessel usually anchored in the lagoon near Runit, approximately 400 m from the shore directly west of the crater. Water samples were routinely obtained from this location before and after the cleanup. On several occasions seawater was sampled in

Table 5. Concentration of radionuclides measured in water from 2 levels in drill hole CD-1; 3/28/80 (Bq m⁻³).

CD-1 depths sampled (m)	^{239 + 240} Pu	S Value	²⁴¹ Am	^{1,37} Cs	90Sr
Solution (<0.45 micron)	2.1	0.097	0.26	10.0×10^{3}	1.1 × 10 ⁴
Particulate (>0.45 micron)	1.6×10^3	0.090	1.6×10^{3}	2.9×10^3	3.0 $\times 10^3$
8.2-9.7					
Solution (<0.45 micron)	1.5	0.068	< 0.1	8.4×10^{3}	1.3×10^{4}
Particulate (>0.45 micron)	$^{4.2}_{\times 10^3}$	0.046	3.3×10^3	8.0×10^{3}	8.5×10^{3}

Table 6. Mean activity of radionuclides measured in soil core samples extracted from the dome and crater by the NAS in 1980.^a

	Bq g 1 dry wt		
Radionuclide	Domeh	Crater ^h	
^{239 + 240} Pu	1.13 ± 0.49	0.35 ± 0.25	
²⁴¹ Am	0.15 ± 0.06	0.23 ± 0.48	
Total TRU (estimate from S value in water)	1.30	0.62	
¹³⁷ Cs	0.34 ± 0.23	0.34 ± 0.34	
⁹⁰ Sr	0.71 ± 0.45	0.81 ± 0.60	

^a Material encased in the donut hole in the center of the dome was not sampled.

the lagoon in the area of the break in the reef north of the crater. A summary of the concentrations of ²³⁹⁺²⁴⁰Pu and ¹³⁷Cs measured during different periods in the filtered seawater samples from these three locations is shown in the lower section of Table 7.

Results in Table 7 show there has been essentially no change in the concentration of ²³⁹⁺²⁴⁰Pu or ¹³⁷Cs in the surface groundwater from the two well sites. Of significance are the results in Fig. 3 showing changes in surface groundwater salinity. Prior to capping the crater, the groundwater (XRU-5 and 6) at these sites was always brackish and had essentially the salinity of seawater. Rainfall impacting on the cement dome results in quantities of freshwater runoff that alters the groundwater quality as is evident from the change in water salinity at CW-1 and 2. Before cleanup, the beach area lagoonward of the crater was barren of vegetation (see Fig. 2a). Now the well sites are overgrown with scrub vegetation and ground cover because of the added fresh water supplied by runoff to the area.

The results shown in the lower section of Table 7 indicate a reduction over time in the amount of dissolved $^{239+240}$ Pu in the offshore surface seawater. The reduction was most notable in the water immediately offshore the crater (CL-1). The $^{239+240}$ Pu concentration in seawater at the station most distant from land was similar to measurements in previous years (CI-1). All concentrations are lower than the levels in the groundwater from the lagoon well sites near the crater that were sampled during comparable times. The water data suggest that the presence of the dome structure has apparently acted to restrict

rather than increase transuranic movement from the crater to the lagoon.

The present mean concentration of ¹³⁷Cs in the lagoon surface water off the crater is many times lower than the groundwater levels in CW-1 and 2 and is now only twice the value of "global fallout" levels found in California Pacific coastal surface waters (Wong et al. 1992) in the late 1980's. It is now comparable to the surface concentrations found in California surface waters during the late 1970's (Noshkin et al. 1978). There has been no evidence of change in the nearshore lagoon seawater concentration of ¹³⁷Cs during the last 16–18 y that could be attributed to any major leakage of from material contained under the dome.

Concentrations in edible flesh of reef fish. The purpose for collections and analysis of specific radionuclides in fish, and in particular plutonium, changed over the years. In the 1970's we were tasked with comparing the levels of radionuclides in tissues of different species of fish from different locations. As the program progressed, dose assessment became the more important issue. This focused our attention on the analysis of the edible flesh from the different species. More than 2,000 fish have been collected from Enewetak Atoll for radionuclide analysis since 1972. Each fish was dissected and the tissue and organs of the species from the same catch were pooled for analysis. It was necessary to pool tissues from a particular catch for analysis because of the low concentrations of transuranic radionuclides encountered in edible muscle tissue and some other parts of the fish.

Differences encountered in the concentration of any radionuclide in the flesh are found to relate to fish species and size; the location where the fish are caught; feeding habits; concentrations in the material ingested; and trophic level. Some of these relationships are demonstrated with the ¹³⁷Cs data in different reef fish from Runit Island shown in Table 8 (Noshkin et al. 1997). The fish described in Table 8 were all caught using throw nets on the lagoon reef near or north of the crater. Data for 2 species of mullet are given along with concentrations in flesh of surgeonfish and goatfish, all used in the local marine diet by Marshallese people. Mullet are herbivorous and detritus feeders. Considerable quantities of bottom sediment are ingested along with food. Adult mullet belong to the 2nd trophic level. Surgeonfish are herbivorous browsers, feeding on algae fronds and fila-

^b Mean value is from 5 samples of soil under the dome and 9 samples below ground level in tremie region.

Table 7. Concentration of radionuclides in filtered surface groundwater from 2 wells lagoonward of the crater and in lagoon surface seawater off-shore North Runit Island. ¹³⁷Cs (d) mean concentration decay corrected to 1/1996. Values in parenthesis are number of samples averaged. Concentrations only in filtered (0.45 micron) surface groundwater or lagoon water.

			Bq m ⁻³	
Years sampled	Groundwater wells	^{2,39 + 240} Pu	¹³⁷ Cs	¹³⁷ Cs(d)
1975–1979	XRU 5 & 6 mean	3.3 (8)	667 (8)	430
(pre disposal)	range	0.4-8.9	74-3,000	
1980-1984	CW 1 & 2 mean	3.2 (16)	890 (10)	640
(post disposal)	range	0.9-9.6	40-2,260	
1994	CW 1 & 2 mean	3.3(2)	555 (2)	560
(post disposal)	range	2.6-4.1	370–740	
	Lag	oon seawater		
	(3 stations 70-	400 m off shore N. Ru	nit)	
1975-1979	mean	2.5 (14)	16 (9)	10
(pre disposal)	range	0.9-4.4	8-22	
1980-1984	mean	1.8 (9)	19 (4)	14
(post disposal)	range	0.6-2.6	10-26	
1994	mean	0.7(3)	10(3)	10
(post disposal)	range	0.7-0.8	6-14	

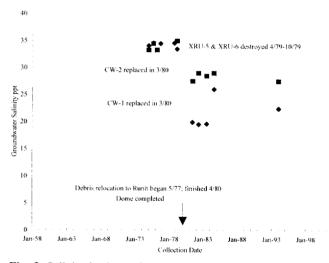


Fig. 3. Salinity in the surface groundwater from 2 wells located lagoonward of the crater.

mentous algae. This species is in the 2nd trophic level. The goatfish consume fossorial as well as surface benthic fauna including small clams, crustaceans, and small benthonic fish and belong to the 3rd trophic level (Noshkin et al. 1981a). The surgeonfish contain higher levels of ¹³⁷Cs in the flesh than either species of mullet or the goatfish. Surgeonfish are more territorial where the mullet and goatfish move in schools to different locations. The concentration in surgeonfish decreases with time, but no such trend is evident in the data for mullet or goatfish. The concentration of ¹³⁷Cs is similar for mullet and goatfish and is essentially below the detection limit in flesh from the fish caught in the 1990's from this region. These differences in muscle concentration relate to the feeding habits of the fish.

Fig. 4 is from Noshkin et al. 1997 and shows a semi-log plot of the data in Table 8 for surgeonfish from North Runit and two additional data points for these fish caught from different locations of the island. One sample was from the ocean reef approximately at mid-island and the other group of surgeonfish was caught in the lagoon from the southern tip of the island. These results demonstrate there are differences related to location sampled even on the same island. The reef fish are territorial and reflect the concentrations in the local environment from which they are caught. The results of repeated sampling of surgeonfish from N. Runit lagoon show a steady rate of decline in concentration with time. This suggests that the data can be used to estimate the change in availability of ¹³⁷Cs to this species from the local environment. The best fit to these data yields a slope related to an effective decay constant of $0.104\pm.012 \text{ y}^{-1}$ with a correlation coefficient of 0.88. The physical half life of ^{137}Cs is 30 y. The computed environmental half-life for ¹³⁷Cs in the lagoon near N. Runit is therefore 8.6 ± 1.0 y. More on the environmental half-life in fish will be discussed in another paper in this volume (Noshkin et al. 1997). The important feature to note is that in recent years the levels in fish indicate no increase that could relate to leakage of

The rate of ²³⁹⁺²⁴⁰Pu disappearance from the environment does not follow that of ¹³⁷Cs because the geochemical behavior of the two radionuclides is significantly different. Between 1972 and 1978, the concentration of global fallout ²³⁹⁺²⁴⁰Pu in surface water of the north equatorial Pacific ocean was equivalent to or less than 0.014 Bq m⁻³ (Noshkin et al. 1987). Table 1 shows that the mean lagoon concentration of ²³⁹⁺²⁴⁰Pu during any year is much greater than fallout background in the Pacific ocean. The difference between the average soluble concentration measured during the different periods indicated is not considered significant, and the assump-

Table 8. Concentration of ¹³⁷Cs in the flesh of four species of reef fish caught in the lagoon off North Runit Island.

	$^{1.57}$ Cs (Bq kg $^{-1}$ wet wt) ^a					
Month/Year collected	Mullet (Neomyxus chaptalii)	Mullet (Crenimugil crenilahis)	Surgeonfish (Acanthurus triostegus)	Goatfish (Mulloidichthys samoensis)		
8/64 ^h			52.0			
11/78		1.0(2)	14.4 (2)			
9/80	0.58 (4)	0.35 (5)		0.64(4)		
9/80	0.26(8)	1.0(3)	7.94(2)	1.41(2)		
7/81	0.52 (12)		9.67 (2)	1.9(7)		
6/82	0.66 (20)		9.11(2)			
8/83	0.67 (18)	1.1 (5)	5.42 (3)			
11/93			8.07 (14)	0.54 (60)		
2/94	1.6 (50)		4.73 (11)			
2/94	1.4 (60)		4.52 (6)			
11/94			1.70 (30)	<1		
5/95	<1		1.83 (35)	< 0.4		
5/95			1.93 (11)	<1		
4/76 Ocean ree	f-mid island		1.6 (5)			
9/80 Lagoon so	outh tip of island		1.7 (3)			

^a Numbers in parenthesis are the 1-sigma counting error expressed as percent of the listed value.

^b Result from Welander et al. (1967).

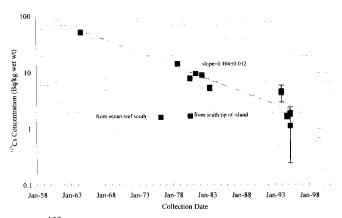


Fig. 4. ¹³⁷Cs concentrations in the flesh of Surgeonfish collected over time from Runit Island.

tion is made that the standing average amount of plutonium mobilized to the lagoon water mass from the Atoll sediments, at any time, is constant. Highest concentrations are generally found in the NE and NW quadrants of the lagoon and lowest levels are generally found in seawater from the SE quadrant. Steady state conditions have been established for ²³⁹⁺²⁴⁰Pu partitioning from the sedimentary reservoir to solution. Between 1972 and 1982, the average "soluble" ²³⁹⁺²⁴⁰Pu was 0.74 Bq m⁻³. The quantity in solution represents only a small fraction of the inventory associated with the sediment. Unlike ¹³⁷Cs, the mobilization of ²³⁹⁺²⁴⁰Pu is a slower process. A similar mean concentration is expected to persist in the lagoon through the remainder of the 1980's and 1990's. Because the physical half-life of ²³⁹⁺²⁴⁰Pu is very long, it is therefore anticipated that the mean concentration of the radionuclide in the flesh of fish from different locations will relate to water levels and will also be relatively constant over time.

Table 9 shows the islands and other locations in the lagoon where reef and pelagic fish were caught since 1972 for plutonium analysis. The islands can be located by referring to Fig. 1. Table 10 shows the ln normal and geometric (median) mean levels of ²³⁹⁺²⁴⁰Pu in flesh of all reef and pelagic fish species collected during different intervals from regions of the lagoon. As with ¹³⁷Cs, there are differences encountered among species with different and similar feeding habits and locations sampled. In general, the level of ²³⁹⁺²⁴⁰Pu found is higher in reef species and lower in pelagic fish. Among the reef fish caught at the same location, the flesh of mullet generally has higher concentrations of plutonium than surgeonfish or goatfish. Fewer samples have been processed for ²⁴¹Am, but the median concentration in samples of flesh from fish from different locations sampled between 1976 and 1982 was 0.0024 Bq kg⁻¹ wet wt. The median concentration best reflects the "average" value in fish but

Table 9. Fish collection locations at Enewetak Atoll.

Island or location ID	Name	US Designator
E-2	Bokombako	Belle
E-9	Boken	Irene
E-10	Enjebi	Janet
E-19	Aomon	Sally
E-20	Bijile	Tilda
E-24	Runit	Yvonne
E-33	Japtan	David
E-35	Medren	Elmer
E-36		Walt
E-37	Enewetak	Fred
E-38	Ikuren	Glenn
E-39	Mut	Henry
E-43	Biken	Leroy
E-45	Drekatimon	Remains-Oscar Tower
E-53		Wide Pass
E-54		Deep Pass

Table 10. Summary of ²³⁹⁺²⁴⁰Pu concentrations (Bq kg⁻¹ wet wt) in the flesh of reef and pelagic fish collected from islands and other locations in Enewetak Atoll.

Fish	from 5 islands (F-2 F	Before cleanup completed (-9, E-10, E-19, E-20) ^a No.		24)
Period	Samples	Number of fish	In n mean	Geometric mean
1972-1978 ^b	21	483	0.22 ± 1.22	0.039
1976–1978	10	464	0.019 ± 0.030	0.010
Fish from Runit (E-24) Period				
1972-1978 ^a	9	129	0.089 ± 0.28	0.027
1976-1978	6	123	0.009 ± 0.006	0.007
Period 1972–1978* 1976–1978	26 6	179 156	$0.074 \pm 0.28 \\ 0.003 \pm 0.004$	0.019 0.002
Fish from 4 islands (E-2	, E-9, E-10, E-19) ^a an	After Cleanup Completed		-24)
Period 1980–1995	27	341	0.007 ± 0.016	0.003
Fish from Runit (E-24)				
Period				
1980–1995	32	819	0.009 ± 0.014	0.005

^a Island names are identified in Table 8 and locations are shown in Fig. 1.

^e No fish were collected from the southern islands after the cleanup because of resettlement.

the In normal mean is shown for information. We tend not to accept some of the data for plutonium generated during the 1972 survey (Nelson and Noshkin 1973) because contamination of the samples is suspected. However, mean values are also included in Table 10 using these data. This is for completeness and comparison if there is disagreement with our assessment on contamination.

Before the cleanup, the median level of ²³⁹⁺²⁴⁰Pu in the flesh of the fish was very low (everywhere only a few mBq kg⁻¹ wet wt). Highest levels were encountered in species from the north islands. The mean concentration was comparable to the average found in all fish from Runit. Lowest levels were measured in fish from the southern part of the Atoll. These observations are supported by the lagoon water concentration data. After the radiological cleanup, no fishing was attempted from the southern part of the Atoll because the Enewetak people were in the process of resettling islands in this region. Fish were collected from Runit and from islands to the north during the early 1980's and 1990's. Again the median concentration of ²³⁹⁺²⁴⁰Pu in the flesh of all fish from both regions during the periods was comparable. The mean in the early 1980's was somewhat lower than the mean computed from the 1993-1995 collections. The 93–95 mean value is comparable to the value in fish from the pre-cleanup years of 1976–1978. There are many explanations for finding a lower mean level in fish during the early 1980's. In the context of this report it is only important to note that there has been no significant change in the mean concentration of plutonium in the

flesh of fish caught in the lagoon near the crater over time. Concentrations are comparable to levels in fish collected from islands on the northern reef of the Atoll during periods before and after the Atoll cleanup exercise. Any TRU's from material at the disposal site have not impacted the marine resources in this region.

SUMMARY

Based on ²³⁹⁺²⁴⁰Pu concentrations measured in soils from regions within the dome after it was filled, the original estimates of the TRU's buried under the dome are questionable. The mean concentrations of the TRU, computed from the core soil sections, are equivalent to or greater than present mean surface soil levels on several northern islands (U.S. DOE 1982). These islands are unrestricted for food gathering. Only 24% of the of transuranics contained under the dome is in communications with the groundwater and available for remobilization. The TRU in the crater is no more than the quantity that has been exposed to seawater for years in the nearshore lagoon sediments off Runit Island. The TRU's in the lagoon are in continuous contact with seawater and are available for uptake by marine fish and other organisms. Levels of ¹³⁷Cs in the flesh of some reef fish from North Runit island are decreasing at a rate faster than radioactive decay alone. No impact is evident from ¹³⁷Cs associated with the debris under the dome. There has been essentially no change in the mean concentration of ²³⁹⁺²⁴⁰Pu in the flesh of reef or pelagic fish over time. The concentrations are comparable to levels measured in

^b Data for flesh in 1972 survey considered suspect and are supplied in this summary for completeness and information only.

the flesh of fish from other regions of the Atoll that are caught and used as part of the marine diet by the Marshallese people. This near constant level in fish is regulated by the slow release and loss of the plutonium from the Atoll sediment reservoir.

Our recent data and conclusions support the findings suggested by the National Academy committee over a decade ago in that any assumption of rapid remobilization of all or any of the dome's transuranics is an extreme one. There is still a presence of TRU's in exposed soils of Runit Island, and for this reason the island has been made off limits. This recommendation should continue to be respected. Quantities of radioactive material in the crater, below ground level, are in communications with seawater and may be mobilized to solution and transported elsewhere. However, levels already present in the sediments and seawater of the lagoon overshadow by orders of magnitude the amounts found under the dome. Concentrations of transuranics in fish are no different now than pre-cleanup levels found in fish from the local environment. Therefore, the present and projected low dose estimates from the marine food chain are little different from those determined in previous years for residents of the Atoll (Robison 1973; Robison et al. 1970; Robison et al. 1987). Any fear that this structure contains amounts of activity whose release would cause damage to the environment that would result in a greater effect on human health is unfounded.

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